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<b>(21) International Application Number:</b> PCT/US99/05238 <b>(22) International Filing Date:</b> 10 March 1999 (10.03.99)  <b>(30) Priority Data:</b> 60/077,733                      12 March 1998 (12.03.98)                      US  <b>(71) Applicant:</b> HYDROGEN BURNER TECHNOLOGY, INC. [US/US]; 3925 Vernon Street, Long Beach, CA 90815 (US).  <b>(72) Inventor:</b> WOODS, Richard, R.; 3925 Vernon Street, Long Beach, CA 90815 (US).  <b>(74) Agent:</b> ABRAHAMS, Colin, P.; Suite 400, 5850 Canoga Avenue, Woodland Hills, CA 91367 (US).		<b>(81) Designated States:</b> AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, UZ, VN, YU, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).  <b>Published</b> <i>Without international search report and to be republished upon receipt of that report.</i>
<b>(54) Title:</b> PROCESS GAS PURIFICATION AND FUEL CELL SYSTEM		
<b>(57) Abstract</b> <p>A module (214) for separating a product from a mixed stream comprises a mixed stream chamber having inlet and outlet means and defining a first flow path for the mixed stream, a purge/product stream chamber having inlet and outlet means and defining a second flow path for a purge/product stream, the second flow path having a substantially countercurrent direction to that of the first flow path, and a membrane located between the mixed stream chamber and the purge/product stream chamber, the membrane being selectively permeable to the product. There is also disclosed a fuel cell system comprising a burner module (210) for mixing and combusting a fuel and air mixture to produce hydrogen rich fuel stream; a hydrogen fuel cell (250) for producing power/energy using the hydrogen fuel produced by the burner module; a hydrogen purification module (214) between the burner module and the fuel cell for extracting hydrogen fuel from the burner module for use in the fuel cell and that uses a purge gas to enhance purification module performance; hydrogen storage means (254) for storing hydrogen fuel produced by the burner module and not immediately required by the fuel cell; and means for feeding stored hydrogen fuel from the storage means to the fuel cell when the hydrogen requirements of the fuel cell are greater than the amount of hydrogen produced in the burner module.</p>		

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PROCESS GAS PURIFICATION AND FUEL CELL SYSTEMField and Background of the Invention

This invention relates to a system of process gas  
5 purification including an apparatus and method for the  
separation of a gas from a mixed gas stream, whereby the  
separated and purified gas can be used in industrial and  
commercial processes. This invention also relates to a system  
whereby the separated gas, for use by a fuel cell, can be stored  
10 in the system for further and later use by the fuel cell.

The invention relates to the purification of a mixed stream  
by separating therefrom a product which forms part of the mixed  
stream. Such purification processes have major industrial  
15 significance, and are also important to small-scale operations.  
The purification process may involve the separation of any one  
of a number of gasses, but more typically pertain to the  
separation of hydrogen or oxygen. These are the two major  
product gases of interest. However, other gases, for example,  
20 nitrogen, argon, carbon dioxide, ammonia, methane and others may  
be separated by the purification process and separation  
apparatus of the present invention.

Conventional purification systems use mixed gas streams  
25 that include the product gas, flowing over a membrane which must  
be permeable to the product gas. On the other side of the  
membrane, the product gas is collected in a pure gas stream and  
allowed to flow out of the system. The effectiveness of these  
conventional systems is very much related to pressure  
30 differentials between the mixed stream gas and the pure gas  
stream that establish the appropriate driving force. Moreover,  
it is critical in conventional systems to ensure a high degree  
of sealing in the system, failing which the pressure  
differentials may result in a flow across the membrane of one or  
35 more gases other than the product gas which may result in  
contamination of the separated product gas. Thus, absolute seals  
between the mixed gas stream side and the pure gas stream side

of the membrane are extremely important. The pressure of the pure product gas stream must be less than the partial pressure of the product gas in the mixed gas stream to ensure a positive driving force toward purification. Because the pressure of the pure product gas stream cannot exceed the partial pressure of the product gas in the mixed gas stream, the pressure of the pure product gas stream must be less than the pressure of the mixed gas stream. Failure of seals or pins holes in the membrane will compromise the purity of the pure product gas stream due to bulk flow of mixed gas into the pure product stream.

Product gas recovery factors, such as the ratio of pure product gas and product gas in the inlet mixed gas stream and the total membrane surface area required may be considered as measures of the effectiveness of the separation process. Usually, the inlet pressure of the mixed gas stream is elevated to several atmospheres, and this helps to minimize the surface area of the membrane in the system, maximize recovery factors and increase the pressure of the pure product gas. As an example of the above, if a pure product gas stream of three atmospheres is needed, the partial pressure of the product gas in the mixed gas stream exit must be greater than three atmospheres. Assuming the mixed gas stream has an inlet product gas concentration of 50%, and a 75% recovery factor was required, the product gas in the outlet of the mixed gas stream would represent one fifth of the gas stream. To illustrate this point, the inlet gas consists of 8 parts, 4 of which are product gas and 4 of which are other gases. Recovery of 75% of the product gas implies that 3 of the 4 product gas parts are removed, and therefore, the remaining mixed gas stream consists of 4 parts other gases and 1 part product gas. The product gas partial pressure at the exit of the mixed gas stream side is  $1/5$  or 20% of the total mixed gas pressure. Since the desired pure product gas is 3 atmospheres, the pressure of the mixed gas stream is 3 atmospheres/20% or equal to 15 atmospheres. Therefore, the mixed gas stream feed pressure would need to be greater than or equal to 15 atmospheres, assuming there was no process flow pressure drop.

The partial pressure driving force across the membrane is 4.5 atmospheres at the inlet (15 atmospheres \* 20%-3 atmospheres), and approximately zero at the outlet. The average driving force is thus 2.25 atmospheres. In such a system, most of the surface area of the membrane is used to achieve the recovery factor, because the driving force approaches zero at the outlet of the mixed gas stream. As a result, the cost and volume of the membrane would be extremely large. Further, the membrane and its seals would have to be constructed so as to maintain cross-pressures of up to 12 atmospheres without leakage in order to maintain the product gas purity.

Some of Applicant's other patents, including U.S. Patent Nos. 5,207,185; 5,299,536; 5,441,546; 5,437,123; 5,529,484; 5,546,701, and well as applications USSN 471,404 and USSN 742,383, are incorporated herein by reference.

#### Summary of the Invention

In one aspect, the present invention is for a gas purification system, wherein a product gas, contained within a mixed gas stream, is removed from the mixed gas stream across a membrane into a purge gas stream flowing counter current to the mixed gas stream on the opposite side of the membrane. The pressures of the mixed gas stream and the purge gas stream respectively are controlled, as is the partial pressure of the product gas on each side of the membrane, to facilitate flow of the product gas through the membrane. The invention also facilitates the use of a system where high precision seals of the membrane and other components, and the total absence of pinholes, is not essential to the purity of the product gas separated from the mixed gas stream. The mixed gas stream and purge gas stream flow counter-current to each other in a separation module. Preferably, the mixed gas stream contains a product gas which is typically hydrogen or oxygen, but the principles of the system can be used to good effect with other product gases, including but not limited to, nitrogen, argon, carbon dioxide, ammonia and methane. Preferably, the purge gas

is an easily separated process gas, and may typically comprise water vapor or a refrigerant, but is not limited to these examples.

5 In one application, the present invention is a method and apparatus for the purification of hydrogen gas from a mixed gas stream exiting from a reformer or an under-oxidized burner.

10 The mixed gas and purge gas streams are separated by an appropriate membrane which must be permeable or effectively permeable to the product gas being separated from the mixed gas stream. To a large extent, the membranes will be selected and incorporated into the separation module according to the nature of the product gas being separated from the mixed gas stream. In  
15 one embodiment, "palladium type" metal membranes have demonstrated effectiveness as hydrogen separation membranes, because the hydrogen gas absorbs into the lattice structure of the metal in direct proportion to the partial pressure of the hydrogen gas. The partial pressure difference between the  
20 hydrogen gas on opposing sides of the membrane is typically used as the driving force to move the hydrogen gas from the mixed gas stream to the purge/product gas stream on the other side of the membrane. Conventionally, these separation membranes operate at elevated temperatures in order to maximize the rate of transfer  
25 of the product gas, in this particular embodiment hydrogen gas is transferred through the membrane. In one embodiment, the product gas transferred is hydrogen.

Other types of membranes that may be used include ceramic  
30 membranes. Ceramic membranes, especially at elevated temperatures, absorb oxygen ions into the lattice structure thereof, and these membranes have therefore been used as oxygen separation membranes. An example of a ceramic membrane is zirconia and yttria stabilized zirconia. Purely ionic conducting  
35 membranes are used in electrochemical reactors, where electric power is the primary driving force for separation. In addition to electrically powered electrochemical reactors, mixed ionic

and electric conducting membranes have been used, where, as with palladium type membranes, only the partial pressure driving force is used to separate oxygen from a mixed gas stream and to purify the oxygen gas.

5

The present invention therefore uses, according to one aspect thereof, an innovative approach to the gas membrane separation process, at the same time reducing or eliminating the problem of ensuring high precision seals and significant pressure differentials which tend to more readily expose any weaknesses in the membrane seals.

In one embodiment of the invention, a separation module permits the flow of two streams through it and separates the streams by an appropriate membrane. On one side of the membrane, a mixed gas stream flows from an inlet to an outlet across the membrane, while on the other side of the membrane, high pressure water vapor is purged in a counter flow direction to that of the mixed gas stream. This inventive separation process maximizes product gas recovery factors and pure product gas pressures, while minimizing the criticality of seals and pinhole-free membranes.

The inventive method for carrying out the purification process, as well as the separation apparatus therefor, has benefits that are clearly illustrated, as will be shown in the following example. A mixed gas stream is fed into the separation module at 15 atmospheres, the mixed gas stream having a 50% concentration of the product gas. The product gas is that component that is to be separated out from the mixed gas stream, and which permeates across the membrane into the purge/product gas stream. In the particular example, a water vapor stream is the purge gas and flows on the opposite side of the membrane to that of the mixed gas stream, and in a counter-flow direction to the mixed gas stream. The volumetric flow rate of the purge gas stream is twice that of the mixed gas stream, and the pressure thereof slightly greater than that of 15 atmospheres. If the

membrane surface area is the same as in the above example, the recovery factor may well approach 100%. Since the partial pressure of the product gas in the purge/product gas is zero at the inlet, the partial pressure of the product gas in the mixed gas stream at the outlet can also approach zero. The partial pressure at the inlet of the purge/product gas stream is in fact zero since the amount of product gas at the inlet of the purge gas stream is zero and, the purge gas at the inlet is pure water vapor.

If all of the product gas is separated or removed from the mixed gas stream, then the purge/product gas stream exiting the module contains four parts product gas and sixteen parts of water vapor. Since the inlet purge gas stream and therefore the purge/product gas stream is at 15 atmospheres, the product gas partial pressure at the exit of the purge/product gas stream exit is approximately 3 atmospheres. The driving force at the inlet of the mixed gas stream is 4.5 atmospheres and zero at the outlet of the mixed gas stream. This translates to an average driving force of 2.25 atmospheres which is the same as the above example and therefore a similar amount of surface area would be needed. The inventive system provides near 100% recovery of the product gas from a mixed gas stream, using similar hardware as conventional systems that achieve only 75% recovery.

In the inventive example described in the preceding paragraphs, the absolute pressure of purge/product gas stream, slightly greater than 15 atmospheres, is higher than the absolute pressure of the mixed gas stream on the other side of the membrane. Any pinholes in the membrane, or poor seals with respect to the membrane or other components of the system, will only allow leakage of purge/product gas into the mixed gas stream, and therefore, the purity of the product gas will not be affected as a result of such leakage. In other words, the absolute pressures are selected such that any leakage which may occur will only flow in the direction that does not compromise the purity of the separated product gas, also minimizing the



need for critical membrane quality, and tightness of the seals within the system.

It is also noted that the cross-pressure, or absolute  
5 pressure differential, is very small, representing only a  
fraction of the absolute pressure. As a result, the membrane  
thickness can be decreased since it will not have to support the  
additional forces resulting from any substantial pressure  
differentials. The ability to decrease the thickness of the  
10 membrane not only enhances the system performance, but also  
reduces the costs thereof.

In another aspect, the invention also relates to the  
treatment of the purge/product gas stream so that product gas  
15 can be delivered to a customer, or downstream equipment, as the  
situation requires. The purge/product gas stream exits the  
separation module at 15 atmospheres, and can thus be delivered  
downstream, at near 15 atmospheres. In one embodiment, this  
delivery may be achieved by including a regenerative steam  
20 generator and water vapor condenser downstream of the separation  
module. At this regenerative steam generator, the water vapor is  
condensed out of the purge/product gas stream, leaving only pure  
product gas at the stream pressure of near 15 atmospheres. In  
turn, the regenerative steam generator is used to recycle as  
25 much of the thermal energy as possible to minimize energy needs.  
In other words, the thermal energy produced by the condensation  
of the water vapor stream is used in the system to heat and  
evaporate water introduced into the separation module at the  
inlet of the purge gas stream. Also this inventive example  
30 illustrates that the pure product gas is at near 15 atmospheres  
or five times the product gas pressure in the conventional  
example.

The separation system and process can be designed so as to  
35 require less surface area, assuming more limited or lower  
recovery factor objectives are acceptable. If, for example, only  
75% recovery is required in the process, the partial pressure of

the product gas at the outlet of the mixed gas stream will be 3 atmospheres, as defined in the original example provided above. The driving force at the mixed gas stream exit is now 3 atmospheres, and therefore the average driving force has increased to 3.75 atmospheres across the entire surface area of the membrane separating the mixed gas stream and the purge gas stream. Thus, the surface area of the membrane needed for the separation is substantially less than when the near 100% recovery was desired. Additionally, the cost and volume of the separation module would also be significantly less since the average driving force is increased and less surface area is required. Nevertheless, even where the recovery requirements are less than near 100%, the overall benefits of the system remain. These include enhanced recovery factors, higher pressure pure product gas exiting from the system, as well as the minimized seal criticality.

#### Brief Description of the Drawings

Figure 1 is a schematic representation of a first embodiment of the present invention illustrating a single membrane component and its inlet and outlet stream;

Figure 2 is a schematic representation of a second embodiment of the present invention illustrating a single electrochemical membrane component, and its inlet and outlet streams;

Figure 3 is a schematic representation of the overall process including a single membrane component and related structures, including a flow circuit of the purge gas stream;

Figure 4(a) is a schematic flow representation showing a conventional fuel cell power system;

Figure 4(b) is a graph showing power loads on the system illustrated in Figure 4(a); and

Figure 5 is a schematic flow representation showing a fuel cell power system of the invention.

#### Detailed Description of the Preferred Embodiments

With reference to the attached drawings, there is shown a

single membrane separation module, both of the ionic and electric power type, as well as a separation module in association with other components. These are shown in Figures 1 to 3 of the drawings. Figures 4 to 5 represent a conventional fuel cell system, and a fuel cell system of the invention, respectively, highlighting the differences and inventive aspects of the fuel cell system as described in this invention.

Reference is now made to Figure 1 of the drawings, which illustrates a separation membrane module, generally designated by reference numeral 10, comprising a gas separation membrane 12 and flow chambers 14 and 16 on each side of the membrane 12. The flow chamber 14 is for receiving and conveying a mixed gas stream 18, while the flow chamber 16 is for receiving and conveying a purge gas stream 20. The mixed gas stream flow chamber 14 has an inlet port 22 at one end and an outlet port 24 at the opposite end. Likewise, the purge gas stream flow chamber 16 has an inlet port 26 at one end and an outlet port 28 at its other end. It will be noted that the inlet port 22 for the mixed gas stream 18 is on the opposite side of the module 10 to the inlet port 26 for the purge gas stream 20, and that the mixed gas stream 18 and purge gas stream 20 therefore flow in opposite directions, or counter current to each other.

The gas separation membrane 12 has a membrane surface 30 which is exposed to the mixed gas stream 18, and a membrane surface 32 which is exposed to the purge gas stream 20. The mixed gas stream 18 at its point of entry into the flow chamber 14 near the inlet port 22 is designated by the reference numeral 34 while the mixed gas stream exiting the mixed flow chamber 14 near the outlet port 12 is designated by the reference numeral 36. With respect to the purge gas stream, the reference numeral 38 represents this stream near the inlet port 26 as it enters the flow chamber 16, while the reference numeral 40 represents the stream 20, including the product, as will be described more fully below, as it exits the flow chamber 16 near the outlet port 28.

In Figure 1, the membrane 12 is preferably a palladium silver type membrane for hydrogen ( $H_2$ ) gas separation. Where the separation module 10 is constructed for the separation of oxygen gas, the membrane 12 may comprise a mixed ionic/electronic conducting ceramic membrane. In any event, it is to be understood that any conventional or other type membrane that is selectively permeable to the desired product gas or liquid, whether hydrogen, oxygen, carbon dioxide, ammonia, methane or other product gas being separated from a mixed gas flow stream, would be appropriate and suitable in the circumstances. The membrane will, of course, be in contact with the respective streams, with the surface 30 of the membrane being in contact with the mixed gas stream, and the surface 32 of the membrane being in contact with the purge gas stream.

Mixed gas stream 34 enters the flow chamber 14 at the inlet port 22. After passing through the flow chamber 14 it then exits as the mixed gas stream 36 through the outlet port 12. This mixed gas stream consists of a stream comprised of the product gas (for example hydrogen, oxygen, or other gas or liquid to be separated from the mixed gas stream) and other gases which are separated from the product gas. On the other side of the separation module 10, a purge gas stream 38 enters through the inlet port 26, travels through the flow chamber 16 and exits as the purge/product gas 40 at the outlet port 28. The purge gas stream can be water vapor or any other gas that can be easily separated from the product gas, as described below, downstream of the separation module 10. Discussed below is a particular embodiment for separating hydrogen or oxygen, as the product gas. Although these gases are discussed in this particular embodiment, other gases or liquids may also be separated from a mixed gas stream within the parameters of the present invention.

Hydrogen is a product gas contained within the mixed gas stream 18 flowing through the flow chamber 14 of the separation module 10. The hydrogen reacts on the surface 30 of the membrane

and is absorbed into the lattice of the membrane 12. The extent of the hydrogen gas in the surface is proportional to the partial pressure of hydrogen in the mixed gas stream above the membrane surface 30. The purge gas stream 20 flows through the flow chamber 16 such that the hydrogen partial pressure at the surface 32 of the membrane is maintained at a partial pressure less than that of the hydrogen at the surface 30 on the other side of the membrane 12. Therefore, hydrogen in the lattice near the surface 30 of the separation membrane 12 migrates through the membrane 12 to the surface 32. Since the partial pressure of the hydrogen near the surface 32 is lower, the hydrogen exits the lattice structure of the membrane 12 and enters the purge gas stream (essentially converting the purge gas stream to a purge/product gas stream) and exits as the stream 40 through the outlet port 28.

The pressure of the purge gas stream 38 near the inlet port 26 and the purge/product gas stream 40 near the outlet port 28, as well as the flow therebetween, is maintained at a pressure greater than the pressure of the mixed gas stream 18. Furthermore, the flow rate of the purge gas stream 20 is maintained sufficiently high to effect a positive hydrogen partial pressure driving force along the entire surface of the gas separation membrane 12. Thus, it will be seen that the effect of maintaining the hydrogen partial pressure higher in the mixed gas stream 18 while maintaining the pressure of the purge gas stream 38 higher in the flow chamber 16 effectively ensures that hydrogen gas can migrate across the separation membrane 12 from the chamber 14 to the chamber 16, but other components of the mixed gas flow stream within the chamber 14 will be prevented from migrating across the separation membrane 12 even if membrane 12 has pinholes or poor seals.

Reference is now made to Figure 2 of the drawings which shows an alternative embodiment of the invention. In this embodiment, the same reference numerals used with respect to Figure 1 have been used for corresponding elements in this

Figure 2. The separation module shown in Figure 2 differs from that in Figure 1 by the presence of an electrode 42 between the flow chamber 16 and the separation membrane 12, and an electrode 44 between the separation membrane 12 and the flow chamber 14.

5 The gas separation membrane 12 illustrated in Figure 2 is an ionic conductive membrane, such as zirconia, but other types of conductive membranes can be used as appropriate.

10 The separation membrane 12 is coated with the electrodes 42 and 44, which are in contact with the purge gas stream 20 and the mixed gas stream 18 respectively. In the example provided below, there is used an oxygen-ion conductive membrane, but other suitable membranes which employ negatively charged ions, or even positively charged ions, are feasible and fall within  
15 the scope of this invention.

Mixed gas stream 18 enters through the inlet port 22 and exits through the outlet port 24 while purge gas stream 20 enters the chamber 16 through the inlet port 26 and exits  
20 through the outlet port 28. Once more, the mixed gas stream 18 and purge gas stream 20 flow counter current to each other. The pressure of the purge gas stream 20 is maintained at a pressure, which is approximately equal to the desired pure product delivery pressure. The pure product, in this case hydrogen, is  
25 delivered downstream of the separation module at a desired pressure, for example 15 atmospheres, and the pressure of the purge gas is maintained at this pure product delivery pressure. At the least, however, the pressure of the purge gas stream 20 must be slightly greater than the mixed gas stream 18 in the  
30 chamber 14 in order to minimize the impact of pinholes or poor seals. If the structure has good seals between the components and the membrane, the pressure criteria can be somewhat less stringent.

35 Electrons 46 enter the electrode 42 and migrate to the reaction site 48 within the electrode 42. In this embodiment, the purge gas stream consists of water vapor, and this water

vapor enters the reaction site 48 with the electrons 46 to form hydrogen, identified by numeral 50, and oxygen ions, identified by numeral 52. The oxygen ion 52 enters the lattice structure of the separation membrane 12. Increased concentrations of oxygens  
5 ions 52 near the reaction site 48 force the oxygen ions 52 to flow to a reaction site 54 contained within the electrode 44. Hydrogen, identified by reference numeral 56, from the mixed gas stream 18 within the chamber 14 enters the reaction site 54 and reacts with the oxygen ions 52. This reaction produces water,  
10 identified by reference numeral 58, and the water vaporizes and enters the mixed gas stream 18 in the chamber 14, at the same time releasing electrons, identified by the reference numeral 60, which leave the electrode 44 and flow through an external circuit 62, ultimately returning to the electrode 42 to complete  
15 the electric circuit.

In the embodiment shown in Figure 2, the driving force to separate the hydrogen is primarily the electrical power, but maintaining a positive partial pressure driving force in  
20 addition to this electrical power can minimize the amount of energy required to drive the system. Further, in this embodiment, and as a result of the electrical driving force, the purge gas flow rate can be decreased because the electrical driving force can move hydrogen from a low partial pressure  
25 mixed gas stream 18 to a higher partial pressure of the purge gas 20 in the chamber 16. Therefore, in this embodiment, unlike that shown in Figure 1, it is not necessary that the partial pressure of the hydrogen within the mixed gas stream 18 is higher than the partial pressure of the hydrogen within the  
30 purge gas stream 20 along the length of the gas separation membrane 12.

The benefits and advantages of the apparatus and method of the invention are equally applicable to other gases. For  
35 example, oxygen in the mixed gas stream could react at the reaction site 54 creating oxygen ions 52 and electrons. The electrons 46 and 60 would therefore have flow directions

opposite to that shown with respect to the situation where hydrogen is being separated from the mixed gas stream, and the oxygen ions 52 would also flow in the opposite direction. At the reaction site 48, the oxygen ions 52 would recombine with the electrons 46 returning from the external circuit 62 to form pure oxygen that would evolve into the purge gas stream, which may typically be comprised of water vapor. The inventive counter-flow purge gas has the same benefits in this case as in the example described above using hydrogen.

Reference is now made to Figure 3 of the drawings, which shows overall a schematic diagram of a gas purification and separation system. In Figure 3, reference numerals used in Figure 1 and 2 have been used for corresponding structures and elements shown in Figure 3. In Figure 3, a mixed gas stream 18 enters through inlet port 22 as stream 34, and becomes a depleted mixed gas stream 36 near the outlet port 24. Purge gas stream 20 enters the inlet 26 as stream 38, and flows in counter-current direction to the mixed gas stream 18. The purge gas 20 exits through the outlet port 28 as the purge/product gas stream 40. The respective flows of the mixed gas stream and the purge gas stream 18 and 20 are isolated by the gas separation membrane 12. This is achieved in accordance with the description above with respect to Figures 1 and 2. The purge/product gas stream 40 flows to a regenerative steam generator 68 where heat is removed from the stream 40 and the water vapor in the stream 40 is condensed. After passing through the steam generator 68, the stream enters a heat removal final stage condenser 70 followed by a liquid/gas separator 72. In the liquid/gas separator 72, the pure product gas stream 76 is isolated from the condensed water, identified by reference numeral 74. The pure product gas stream 76 is removed from the stream for use or delivery to a downstream site.

The water 74 flows through a pump 78 and back into the regenerative steam generator 68, where the liquid water 74 is converted to water vapor by heat transferred from condensing



water from the stream 40. If a refrigerant type purge gas is used, an orifice 80 is placed between the pump 78 and the steam generator 68. The water vapor is further heated in a super heater 82 and becomes the purge gas stream 38 that enters the inlet port 26 of the separation module 10. Additional water may be added to the separator 72, if needed.

Reference is now made to Figures 4(a), 4(b) and 5 of the drawings. Figure 4 shows a conventional fuel cell power system, while Figure 5 shows the fuel cell system of the invention.

In Figure 4(a), there is shown an under-oxidized burner/reformer, or fuel processor, 110. The fuel processor 110 receives, for example, hydrocarbon fuels such as methanol, ethanol, diesel and the like, mixes and combusts them so as to produce a hydrogen rich mixed gas product stream. The hydrocarbon fuel is introduced through fuel inlet 112, and is conveyed along the fuel line 114, assisted by pump 116. A valve 118 is located in the fuel line 114 so as to control the flow of fuel therethrough. The fuel line 114 enters the fuel processor 110. An air inlet 120 is provided in a turbo generator 122 and the air is forced along air line 124 to the fuel processor 110. Water vapor may be incorporated into the air from line 154. In the fuel processor 110, the air from air line 124 and the fuel from fuel line 114 are combined, reacted and combusted, preferably in the presence of water, to produce the hydrogen/mixed gas product which exits the fuel processor 110 through the product line 126. The product line 126 conveys the hydrogen/mixed gas product through downstream shift reactors 170 and 172, heat recovery heat exchangers 173 and 175, and through carbon monoxide polishing unit 176 and eventually to a fuel cell 128 where it is combined with air pumped through air line 130, the air in air line 130 having as its source the turbo generator 122.

The fuel cell 128 has an anode exhaust line 132 and a cathode exhaust line 134, each of which connects to a condenser

136 and 138 respectively. The condenser 136 connects to a separator 140 while the condenser 138 connects to separator 142, the separators 140 and 142 extracting water from the mixture, with the water being drained off through water lines 144 and 146 respectively. The water lines 144 and 146 consolidate into a single water line 148, which conveys the water to steam generator 150. Some of the water, however, is tapped off through water line 152 for introduction into the fuel processor 110, where it is mixed with the hot product gas from the combustion chamber. The water reaching the steam generator 150 through the consolidated water line 148 is heated by recovered heat from heat exchangers 173 and 175, and resulting water vapor is supplied through water vapor line 154 to the air line 124, where air and water vapor mix and are subsequently introduced to the fuel processor 110, as described above.

Each separator 140 and 142 is connected to a combustor 156 by product residue lines 158 and 160, and the product residues are burned in the combustor 156 to produce heat and energy which flow through the line 162 and feeds the turbo generator 122. The heat and energy contained within the product flowing through line 162 is processed by the turbo generator and is used to power a generator 164. The combustor gas is exhausted through the exhaust line 166 after the turbo generator 122.

In the fuel processor 110, there is a combustion chamber 168 where the air, fuel and water mixture are combusted, and where temperatures reach approximately 2700°F. In the lower portion of the fuel processor, water from line 152 is introduced to quench the product gas down to approximately 700°F. Downstream there is provided a high temperature shift region 170 and a low temperature shift region 172, where carbon monoxide is reacted with water to form additional hydrogen and carbon dioxide. These shift reactors assist in ridding the system of undesired byproducts of combustion. The fuel processor process further comprises a zinc oxide bed 174 where sulfur is extracted from the combusted mixture. The product gas exiting the sulfur

bed 174 and high temperature shift 170 is cooled by heat exchanger 173 in preparation for entering low temperature shift reactor 172. The gas is also cooled, after the low temperature shift reactor 172, by heat exchanger 171. Finally, the carbon monoxide is reduced in the carbon monoxide polishing unit 176 to levels acceptable to the fuel cell 128. The hydrogen/mixed gas product in the product line 126 has a fairly low concentration of hydrogen, and may typically comprise only 30% to 40% of the total product gas.

It has been described above that the combustor 156 receives residual products from the fuel cell 128 via the condensers and separators, and these are combusted so that the product traveling through line 162 are at elevated temperatures. The temperature of these products in line 162 may reach approximately 800°F, significantly lower than temperatures reached in corresponding structures of the fuel cell system of the invention, as will be described below.

Reference is now made to Figure 5 of the drawings, which is a diagrammatic flow chart illustrating the components and system of the invention. As will become apparent from the discussion below, the fuel cell system of the invention offers many advantages for increased efficiency and output, requiring an under-oxidized burner having a nominal or lower power rating than in conventional systems. The acceptability of the lower power rating is due to the capacity of the system of the invention to use and produce hydrogen fuels with greater efficiency, and to store hydrogen in a reservoir for subsequent use. With the ability to store hydrogen, the under-oxidized burner producing the hydrogen need not operate at erratic high peak power requirements, but can typically operate at more consistent and steady states yet still provide sufficient hydrogen to enable the system to operate at peak power levels of conventional systems.

With reference to Figure 5, there is shown an under-

oxidized burner/reformer 210 including, generally, a combustion chamber 212. The inventive system integrates an innovative purification module 214 downstream of the combustion chamber 212. The purification module 214 has a mixed gas side 213 and a  
5 purge/product gas side 233. The mixed gas side 213 flows in counter flow direction to the purge/product gas side 233. The under-oxidized burner 210 is supplied with air, fuel and water which are ignited and thoroughly mixed to produce hydrogen, carbon monoxide and water.

10

A fuel inlet 216 is provided and supplies a fuel line 218 which is pumped by a pump-compressor 220 to the combustion chamber 212. A valve 222 is provided for controlling the flow of fuel through the fuel line 218. As with conventional systems,  
15 the fuel may comprise hydrocarbon fuels, such as methanol, ethanol, diesel or other appropriate fuels. An air inlet 224 provides air to a turbo generator 226 and air line 228 conveys the air from the turbo generator 226 to the combustion chamber 212 of the burner 210. The air flowing through air line 228 may  
20 be supplemented with steam, provided by steam line 230 emanating from another source within the fuel cell system, as will be described further below. The steam line 230 has a branch line 232 which provides steam to the purification module 214.

25

The burner 210 has located within the combustion chamber 212 structures for thoroughly mixing the air and fuel, received through lines 228 and 218 respectively. These structures typically terminate in an injector nozzle where a thoroughly mixed fuel and air mixture is discharged into the combustion  
30 chamber for ignition. The mixing structures and injector nozzles are fully described in some of Applicant's other patents, including U.S. Patent Nos. 5,207,185; 5,299,536; 5,441,546; 5,437,123; 5,529,484; 5,546,701, and well as applications USSN 471,404 and USSN 742,383, all of which are incorporated herein  
35 by reference.

After the combustion process within the burner

210 has converted the hydrocarbon fuels into hydrogen and carbon monoxide mixed gas stream, the mixed gas stream enters the purification module 214 and flows through the mixed gas side 213. Steam from line 232 enters the purification module 214 and flows through purge/product gas side 233. The mixed gas flow 213 and purge/product gas flow 233 flow in counter flow relationship to each other. Hydrogen from the mixed gas flow 213 is transferred into the purge/product gas flow 233 as described for Figures 1 and 2. Recovery factors of 70 to 90% are preferred.

The hydrogen/steam mixture exits from the burner 210 through the water/hydrogen line 236 where it is conveyed to a condenser 238 which can also be a recuperative condenser as described in Figure 3. A final condenser 240 is provided, the condenser 238 and final condenser 240 constituting a water condenser train. The condensers convert the hydrogen/steam into hydrogen/liquid water mixture and a separator 242 is provided for separating and isolating these two components. The hydrogen exits the separator 242 through the hydrogen line 244, while the water exits the separator 242 through a water line 246.

A particular advantage of the invention is that the hydrogen line 244 contains near 100% hydrogen gas, as compared with conventional systems where only 30% to 40% of hydrogen is contained in a hydrogen-product mixture fed to a fuel cell. In the present invention, the hydrogen line 244 can be directed either through line 248 to a fuel cell module 250, or through line 252 to a hydrogen storage tank 254. Clearly, the amount of hydrogen gas entering either one of lines 248 and 252 respectively will depend upon the load exerted on the fuel cell 250. If all the hydrogen gas from the separator 242 is needed to meet current load requirements of the fuel cell 250, all of the hydrogen produced will go through the line 248 to the fuel cell 250. On the other hand, if the hydrogen emanating from the separator through line 244 exceeds current load requirements, all or part of the hydrogen will flow through line 252 for storage in the hydrogen storage tank 254. A pump 256 is provided

in line 252 for transmitting the hydrogen gas to the storage tank 254.

It is, of course, quite possible that the load on the fuel cell 250 will be such that greater amounts of hydrogen will be required than are in fact being produced and separated within the separator 242. In conventional systems, these increased requirements would place strain on the fuel processor to produce more hydrogen. In the present invention, however, the additional hydrogen requirements are met by drawing on the reservoir of hydrogen within the storage tank 254. Line 252, the flow of gas through which is controlled by valve 258, provides a passage whereby hydrogen can be fed from the storage tank into the line 248 for use by the fuel cell 250.

A cathode exhaust line 260 is provided and conveys the cathode exhaust products from the fuel cell to a catalytic combustor 262. The catalytic combustor 262 is also provided, through feed line 264, with depleted gas products from the burner 210. These depleted gas products from the mixed gas side 213 of the purification module include hydrogen gas. Furthermore, the catalytic combustor 262 is provided with compressed air through line 266 from the turbo generator 226. The catalytic combustor 262 burns the products received from the burner 210, namely the depleted gas, as well as the cathode exhaust products provided from the fuel cell through line 260. Air provided through line 266, the flow whereof is controlled by valve 268, enhances the combustion process, which results in the production of significant amounts of heat. The catalytic combustor product gases exit through line 270, at temperatures typically ranging between 1200°F and 1800°F which is, once more, a substantially higher temperature than that produced by conventional systems. As will be recalled with reference to Figure 4(a), the combustor burns products and raises temperatures to approximately 800°F. The line 270 supplies the turbo generator 226, and the energy, at least in part, is used to drive the generator 272. The line 270, conveying products

under pressure, is transmitted to the expander portion 226a of the turbo-generator and the pressure and heat drives the expander. The expander outlet gas flows through line 291 to recuperative steam generator 284, condenser 288, and liquid/gas separator 278. Water from separator 278 is combined with water from separator 242 and enters pump 286. Water from the pump 286 flows in the line 282 through control valve 289 to steam generator 284. The steam is then sent to line 230.

As will be noted from Figure 5, the energy, at least in part, used for converting the water to steam in the steam generator 284 is derived from the exhaust product from the turbo-generator 226, which, unlike, conventional systems, is not discharged from the system, but is conveyed through line 291 to the steam generator 284. If additional energy is needed, the recovery energy from condenser 238 can be integrated with the heat supplied by line 291 to generator 284. This excess heat and energy is thus used in the inventive system, compared to conventional equipment where it is discharged. The present system is thus more efficient in utilizing all energy sources, resulting in less waste and lower fuel requirements for the amount of energy produced.

The steam produced in the steam generator 284 exits through steam line 230, and is fed into the air line 228 where the air and steam combination is transported to the burner 210 and fuel cell 250. A steam branch line 232 is provided whereby steam from the steam generator 284 is introduced to the inventive purification module 214.

The system of the invention has several significant advantages compared to conventional fuel cell systems. One difference relates to the start up times of conventional systems versus the system of the invention. In conventional systems, start up requirements may take at least 2 minutes before hydrogen production is increased to meet the load requirements placed on the fuel cell. In practice, a typical profile of

energy use is highly erratic, with demand for power alternating between high and low points. Figure 4(b) of the drawings shows a graph of typical profile uses of energy in conventional systems. The system surges as it runs, as load requirements increase or decrease, resulting in higher production of hydrogen. The system must be designed to cope with such surging, and to deal with the great variations in demand. The system of the invention, of course, incorporates a hydrogen storage tank 254. Therefore, start up times are significantly reduced since the hydrogen from the storage is readily available for immediate use by the fuel cell. This hydrogen storage tank is supplied with hydrogen when the fuel cell load requirements call for less hydrogen than is being produced by the burner. Conversely, during surging, additional hydrogen required to power the fuel cell is obtained not from the higher output of the fuel processor, but rather from the fuel storage tank.

The ability of the inventive system to draw on a hydrogen reserve during peak power surges facilitates another particularly advantageous feature of the invention, namely, that the fuel processor can be designed to put out less hydrogen at its peak performance. Since conventional systems with 30 to 40% hydrogen streams have not had the ability to store hydrogen, it has been necessary to provide a fuel processor which is able to output the necessary hydrogen at levels corresponding generally to the peak power requirements. In fact, peak power demands can, in typical systems, be 50kW or more even though the average load may only be 15kW. The net effect of the ability to store hydrogen is that, instead of demands being made on the fuel cell and reformed, the system resorts to stored hydrogen to compensate for the lower levels of production during peak demand. The system of the invention therefore permits the fairly regular and consistent output of hydrogen from its fuel processor, whether or not it is needed. For that hydrogen which is not immediately needed, the hydrogen storage tank comes into operation and constitutes a reservoir of the hydrogen to be drawn upon during peak demand when requirements exceed the



output capacity of the fuel processor. Therefore, the fuel processor and/or reformer can be appropriately sized to produce a base load. Apparatus able to produce 15kW instead 50kW can be used not only lowering the cost of the overall system, but also making the net amount of hydrogen produced for a given amount of fuel much more efficient. The actual size of the under-oxidized burner and reformer can also be reduced influencing space saving factors. The burner and fuel cell can therefore operate consistently at near maximum efficiency, at the same time having some reserve capacity, due to the ability to store excess hydrogen gas which is not used. With the ability to store hydrogen, the fuel processor can be designed to produce hydrogen at steady levels, allowing structures which are 30% or less than the size of fuel cells and/or reformers which need be used in conventional systems.

As will be apparent from the description above, the system of the invention is also superior to conventional systems in that it produces substantially higher concentrations of hydrogen. In the conventional system illustrated in Figure 4(a), the hydrogen/mixed gas in product line 126 contains hydrogen concentrations comprising 30-40% of the mixture. In comparison, hydrogen exiting the burner 210 shown in Figure 5, through the hydrogen line 244 is near 100% of the mixture. It is extremely difficult to efficiently store the hydrogen/mixed gas combination, where hydrogen concentration is as low as 30-40%. This problem does not arise in the system of the present invention, owing to the near purity of the hydrogen within the mixture exiting the burner. The high purity hydrogen also improves the effectiveness of the fuel cell 250 which results in smaller, lower cost fuel cell stack.

The system of the invention also has the ability to operate the turbo generator at higher temperatures, increasing its efficiency of operation. In Figure 4, the fuel processor 110 used in conventional systems must of necessity include certain shift processors in the combustion apparatus in order to get rid

of undesirable components such as carbon monoxide. These shift reactors result in a substantial reduction in the thermal content, particularly from the high temperature shift region to the low temperature shift region in the fuel processor. The system of the invention separates the hydrogen gases from the remainder or depleted gas. The hot depleted gas from the purification module 214 is sent directly to the catalytic combustor 262 to produce heat.

10 In the present invention, the system allows temperatures as high as 1200° - 1800°F in this context, which not only feeds the expander and generator, but also results in a residual heat and energy which is not exhausted, but recycled to a heat recovery steam generator where water in the system is converted to steam for further use in the hydrogen production process. Hence, turbo-generator performance increases and the system puts out more energy for each unit of fuel.

20 The system of the invention also reduces the complexity of the fuel processing system and the fuel cell system. In conventional fuel processing systems, shift reactors 170 and 172, carbon monoxide polishing unit 176, and sulfur absorbent bed 174 are required to eliminate contaminants in the product gas entering the fuel cell. The mixed gas side of the purification module 214 can be designed to integrate shift catalysts functions and therefore, the shift reactors 170 and 172 are not required. The function is enhanced because of the removal of hydrogen through the purification module 214. Since the reformer product gas does not directly pass through the fuel cell module 250 the sulfur absorbent bed 174 and carbon monoxide polishing bed 176 are also not required. The elimination of these components also reduces the system size and cost.

35 As described in further detail with respect to Figures 1 to 3, steam purging in the separation module is able to provide greater than 85% recovery of hydrogen gas within the mixed stream, supply hydrogen gas to the fuel cell at high pressure,

and, as a result of the pressure differentials on each side of the membrane within the module, minimize any adverse effects resulting from pinholes or poor seals, thus making these seals less critical.

5

With the reservoir of hydrogen gas, stored from hydrogen production from the fuel cell when it produces more hydrogen than is required by the system, a rapid acceleration and start up time is facilitated, since drawing on a reservoir of stored  
10 hydrogen takes far less time to achieve than accelerating the fuel cell to surge and provide increased outputs of hydrogen. The invention does not require shift catalyst beds and/or sulphur removal beds, necessary in conventional systems to deal with the fuel processes production of undesirable components.  
15 Since these undesirable components are not produced in significant quantities, higher temperatures can be maintained within the system, increasing efficiency.

A typical start up cycle of the fuel cell system of the  
20 invention involves a battery spin of the turbo generator to start air flow to the fuel cell and hydrogen is released from the storage tank 254. The time that is required for this start is very short. Air flows to the under-oxidized burner 210 and catalytic combustor 262, which are thermal processes, and  
25 therefore, typically require longer, more gradual start up cycles. The fuel flow to the under-oxidized burner 210 starts, and a spark plug within the burner is activated to ignite the mixture within the combustion chamber 212. The burner 210 is operated at a high capacity and high stoichiometric (SR) ratios.

30

The combination of heat to the turbo generator 226 and hydrogen from the storage tank 254 to the fuel cell 250 provides instant vehicle movement, or the instant necessary power output for the particular application. As the system heats up, steam is  
35 formed providing purge gas through line 233 to the hydrogen concentration module, and the system reaches an operating temperature so as to extract hydrogen from the mixed gas stream

213. The purification module 214 begins to extract hydrogen from the mixed gas stream 213 and begins to provide hydrogen to the fuel cell 250. As the fuel cell 250 load decreases, some hydrogen begins to be added to the storage tank 254. The hydrogen storage cycle, wherein hydrogen is diverted to the storage tank, recharges the hydrogen storage tank, as requirements demand.

The process gas purification module and the fuel cell system, provide effective and efficient utilization of hydrogen from hydrocarbon fuels. The under-oxidized burner of the invention, in association with the gas purification module, is designed so as to extract optimal quantities of hydrogen from the hydrocarbon fuels. This is achieved by controlling the partial pressure of hydrogen (or other gas being extracted from a mixed gas stream), on each side of the membrane in a purification module, and by manipulating the overall pressure and partial pressures of the streams flowing on each side of the membrane. Furthermore, the invention goes one step further in that, not only are optimal hydrogen concentrations extracted from the hydrocarbon fuels, but also the actual utilization of the hydrogen following this process is optimized. Higher temperatures and pressures are used more effectively for delivery of hydrogen to the fuel cell and for the production of electricity. Moreover, the fuel processor can be smaller, and operated at more consistent levels since fuel cell loads draw not only on the hydrogen production from the fuel processor, but also on a storage tank which is filled and recharged during lower load periods.

30

The invention is not limited to the precise details described hereabove, and a number of alternative embodiments may be used within the scope of the invention. Central to all these embodiments is the use of a purge gas on the product gas side that flows in a counter flow direction to the mixed gas stream.

35

CLAIMS:

1. A module for separating a product from a mixed stream, the module comprising:

5 (a) a mixed stream chamber having inlet and outlet means and defining a first flow path for the mixed stream;

(b) a purge/product stream chamber having inlet and outlet means and defining a second flow path for a purge/product stream, the second flow path having a substantially countercurrent direction to that of the first flow path;

10 (c) a membrane located between the mixed stream chamber and the purge/product stream chamber, the membrane being selectively permeable to the product.

15 2. A module as claimed in claim 1 wherein the purge/product stream chamber is connected to a source of purge gas.

20 3. A module as claimed in claim 1 wherein the purge/product stream chamber is connected to a source of supply for providing the purge/gas stream.

4. A module as claimed in claim 1 wherein the inlet means to the purge/product stream is connected to a source of condensable gas, the condensable gas being selected from the group consisting of high pressure water vapor, alcohol vapors, 25 fluorocarbon vapors, chloro-fluorocarbon vapors and any refrigerant type compound.

30 5. A module as claimed in claim 1 wherein the mixed gas chamber is connected at its inlet means to a partial oxidation reformer, and the product to be separated from the mixed stream is hydrogen.

35 6. A module as claimed in claim 1 wherein the outlet means for the purge/product stream chamber is connected to a purge gas condenser downstream thereof to separate the product from the purge/product stream.

7. A module as claimed in claim 1 wherein the membrane is a palladium type membrane that is permeable to hydrogen gas.

8. A module as claimed in claim 1 wherein the membrane comprises a first surface exposed in the mixed stream chamber, a second surface exposed in the purge/product stream chamber and a lattice structure between the first and second surfaces of the membrane selectively permeable to the product.

9. A module as claimed in claim 1, further comprising a first electrode on a surface of the membrane exposed in the mixed stream chamber and a second electrode on a surface of the membrane exposed in the purge/product stream chamber, and connecting means between the first and second electrodes for the flow of electrons.

10. A module as claimed in claim 9 wherein the membrane is a negative ion conducting membrane.

11. A module as claimed in claim 9 wherein the negative ion conducting membrane is an oxygen ion conducting membrane.

12. A module as claimed in claim 11 wherein the oxygen ion conducting membrane is zirconia.

13. A module as claimed in claim 9 wherein the connecting means is an external circuit whereby free electrons are conducted outside the module through the external circuit from the first to the second electrodes.

14. A module as claimed in claim 9 wherein the membrane is an oxygen ion conducting membrane, the oxygen ion conducting membrane comprising a mixed ion and electron conducting material, with no external circuit.

15. A module as claimed in claim 9 wherein the membrane is an oxygen conducting membrane and is comprised of a mixed ion and

electron conducting material, and the connecting means is an electrical connection to an external circuit for the flow of free electrons.

5 16. A module as claimed in claim 1 wherein the mixed stream chamber further comprises a catalyst to promote additional reactions.

10 17. A module as claimed in claimed 16 wherein the catalyst promotes a carbon monoxide shift reaction.

18. A module as claimed in claim 1 further comprising a downstream condenser.

15 19. A module as claimed in claim 1 further comprising a water separator for condensing out water vapor from the purge/product stream, and means for conveying the condensed water to the inlet means of the purge/product stream chamber, the condensed water being heated by a steam generator and superheater prior to inlet  
20 into the purge/product stream chamber.

20. A method of treating a mixed stream by separating therefrom a product, the method comprising:

25 (a) introducing the mixed stream having a first flow path into a mixed stream chamber of a module, the mixed stream chamber having inlet and outlet means,

(b) introducing a purge stream having a second flow path into a purge/product stream chamber of the module, the purge/product stream chamber having inlet and outlet means such  
30 that the second flow path is substantially counter-current to the first flow path,

(c) locating a separation membrane between the mixed stream chamber and the purge/product stream chamber, the separation membrane being selectively permeable to the product such that  
35 the product evolves from the mixed stream through the separation membrane and into the purge stream, to thereby form a purge/product stream,

(d) removing the purge/product stream from the purge/product stream chamber through the outlet means thereof, and

5 (e) removing the product depleted mixed stream from the mixed stream chamber through the outlet means thereof.

21. The method of claim 20 wherein the partial pressure of the product at a point on the separation membrane in the mixed stream chamber is greater than the partial pressure of the  
10 product at the point on the opposite side of the separation membrane by way of providing a purge gas through the product side of the membrane, thereby providing a driving force for the product to permeate the separation membrane.

15 22. The method of claim 20 wherein the pressure of the mixed stream in the mixed stream chamber is less than the pressure of the purge stream and the purge/product stream in the purge product stream chamber.

20 23. The method of claim 20 wherein a driving force for the product to permeate across the separation membrane comprises electrochemical reactions on opposite sides of the separation membrane.

25 24. The method of claim 21 wherein a driving force for the product to permeate across the separation membrane further comprises electrochemical reactions on opposite sides of the separation membrane.

30 25. The method of claim 20 wherein the purge stream is water vapor or steam.

26. The method of claim 20 wherein the separation membrane is active at elevated temperatures.

35 27. The method of claim 26 wherein the elevated temperatures are greater than 400°F.



28. The method of claim 20 wherein the purge stream is a non-reactive vapor having a relatively high vapor pressure at operating temperatures and a relatively high condensation temperature to facilitate separation thereof from the product.

29. The method of claim 20 wherein the purge gas is selected from the group consisting of an alcohol, a fluorocarbon, and any refrigerant type compound.

30. The method of claim 20 wherein the product is a gas which is selected from the group consisting of: hydrogen, oxygen, nitrogen, argon, carbon dioxide, ammonia and methane.

31. A fuel cell system comprising:

(a) a burner module for mixing and combusting a fuel and air mixture to produce hydrogen rich fuel stream;

(b) a hydrogen fuel cell for producing power/energy using the hydrogen fuel produced by the burner module; and

(c) a hydrogen purification module between the burner module and the fuel cell for extracting hydrogen fuel from the burner module for use in the fuel cell and that uses a purge gas to enhance purification module performance.

32. A fuel cell system as described in claim 31 further comprising:

(a) hydrogen storage means for storing hydrogen fuel produced by the burner module and not immediately required by the fuel cell;

(b) means for feeding stored hydrogen fuel from the storage means to the fuel cell when the hydrogen requirements of the fuel cell are greater than the amount of hydrogen produced in the burner module.

33. A system as claimed in claim 31 wherein the burner module comprises an under-oxidized burner for producing at high temperatures a hydrogen rich fuel stream mixture.

34. A system as claimed in claim 32 further comprising  
condensing means between the burner and the fuel cell for  
extracting water vapor from the hydrogen fuel and water vapor  
5 mixture, the hydrogen fuel being conveyed to the fuel cell  
and/or storage tank, as required.

35. A system as claimed in claim 31 further comprising a  
combuster for combusting depleted gas produced by the burner,  
10 the combuster yielding additional heat and energy for driving a  
generator.

36. A system as claimed in claim 35 wherein the combuster is  
catalytic combuster.

15 37. A system as claimed in claim 35 further comprising a heat  
recovery steam generator, the steam generator being driven by  
the additional heat and energy produced by the combuster and  
heat recovered by the condensers.

20 38. A system as claimed in claim 31 further comprising a turbo  
generator for providing air under pressure to the burner.

39. A method of providing hydrogen to a fuel cell, the method  
25 comprising:

(a) mixing and combusting a fuel and air mixture in a  
burner module to produce hydrogen rich fuel stream;

(b) providing a hydrogen fuel cell for producing  
power/energy using the hydrogen fuel produced by the burner  
30 module;

(c) providing a hydrogen purification module between the  
burner module and the fuel cell for extracting hydrogen fuel  
from the burner module hydrogen rich stream to the fuel cell;

(d) storing hydrogen fuel produced by the burner module  
35 and not immediately required by the fuel cell in a hydrogen  
storage means, and;

(e) feeding stored hydrogen fuel from the storage means to

the fuel cell when the hydrogen requirements of the fuel cell are greater than the amount of hydrogen produced in the burner module.

5 40. A method as claimed in claim 39 further comprising the step of purging water vapor through the purification module to enhance separating out hydrogen fuel from the hydrogen rich stream so as to provide substantially 100% hydrogen fuel to the fuel cell and storage means.

10

41. A method as claimed in claim 39 wherein hydrogen is provided to the fuel cell at elevated pressures.

15 42. A method as claimed in claim 39 wherein depleted gas produced by the burner is ignited in a catalytic combustor to produce additional heat and energy, the heat and energy driving a generator.

20 43. A method as claimed in claim 42 wherein the additional heat and energy is also conveyed to a heat recovery steam generator, the steam generator heating water to steam for use in the burner and purification module.

25 44. A method as claimed in claim 43 wherein the water heated to steam comprises water separated from the hydrogen fuel water vapor mixture produced by the purge gas enhanced purification module.

30 45. A method of extracting hydrogen from a hydrogen rich stream comprising:

(a) passing a hydrogen rich stream on one side of an oxygen conducting ceramic membrane;

(b) passing water vapor on the other side of the oxygen conducting ceramic membrane;

35 (c) promoting the reaction of hydrogen from the hydrogen rich stream with oxygen ions in the membrane to form water vapor; and

(d) promoting the reaction of water vapor on the other side of the membrane to form hydrogen and oxygen ions.

46. The method defined in claim 45 further comprising:

- 5       (a) the electrochemical reaction of the hydrogen and oxygen ions on the mixed gas side of the membrane; and
- (b) the electrochemical reaction of the water vapor on the other side of the membrane to form hydrogen and oxygen ions.

10

FIG. 1

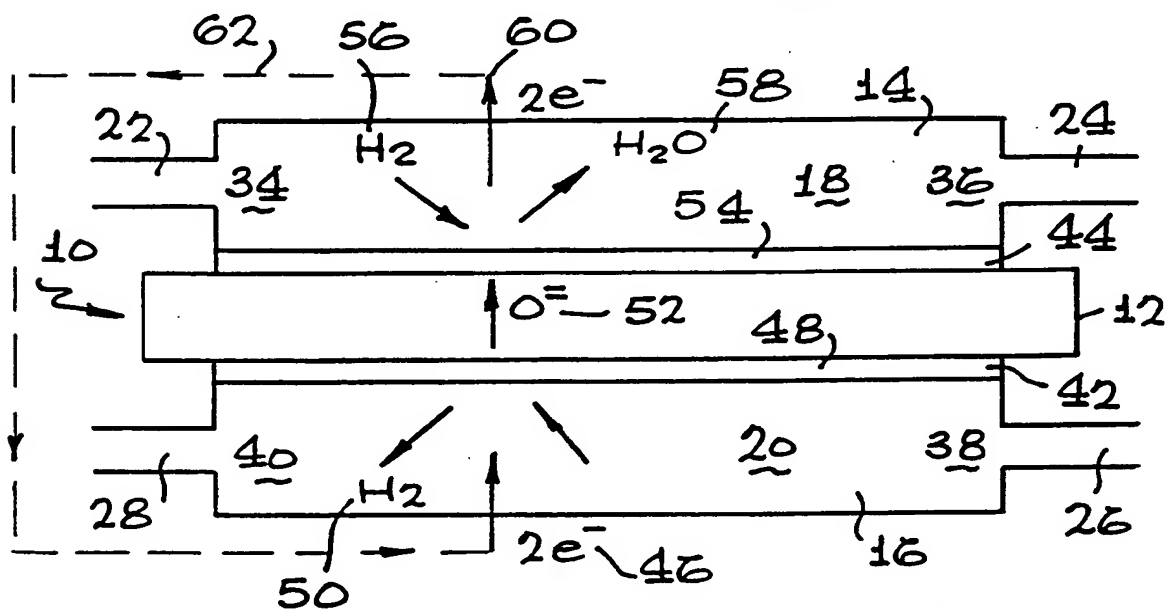
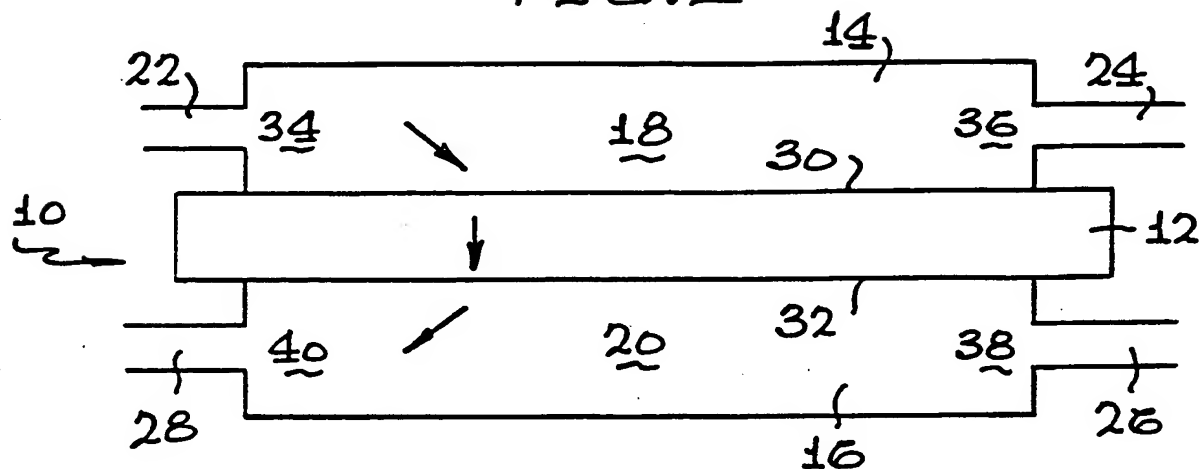


FIG. 2

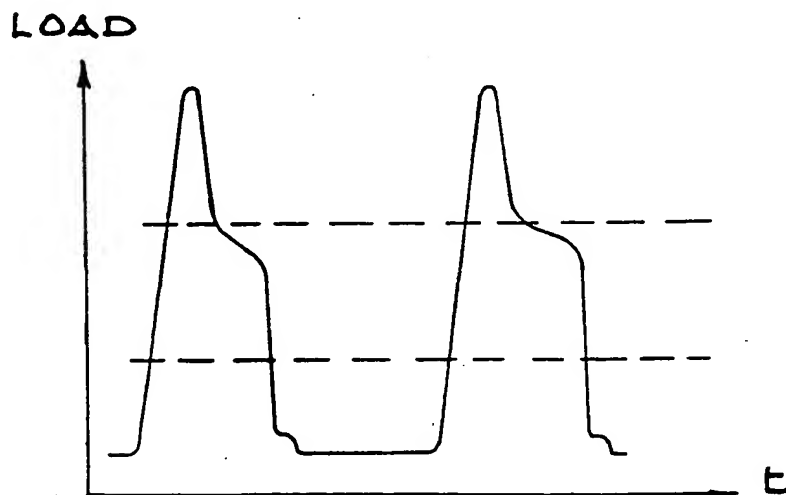
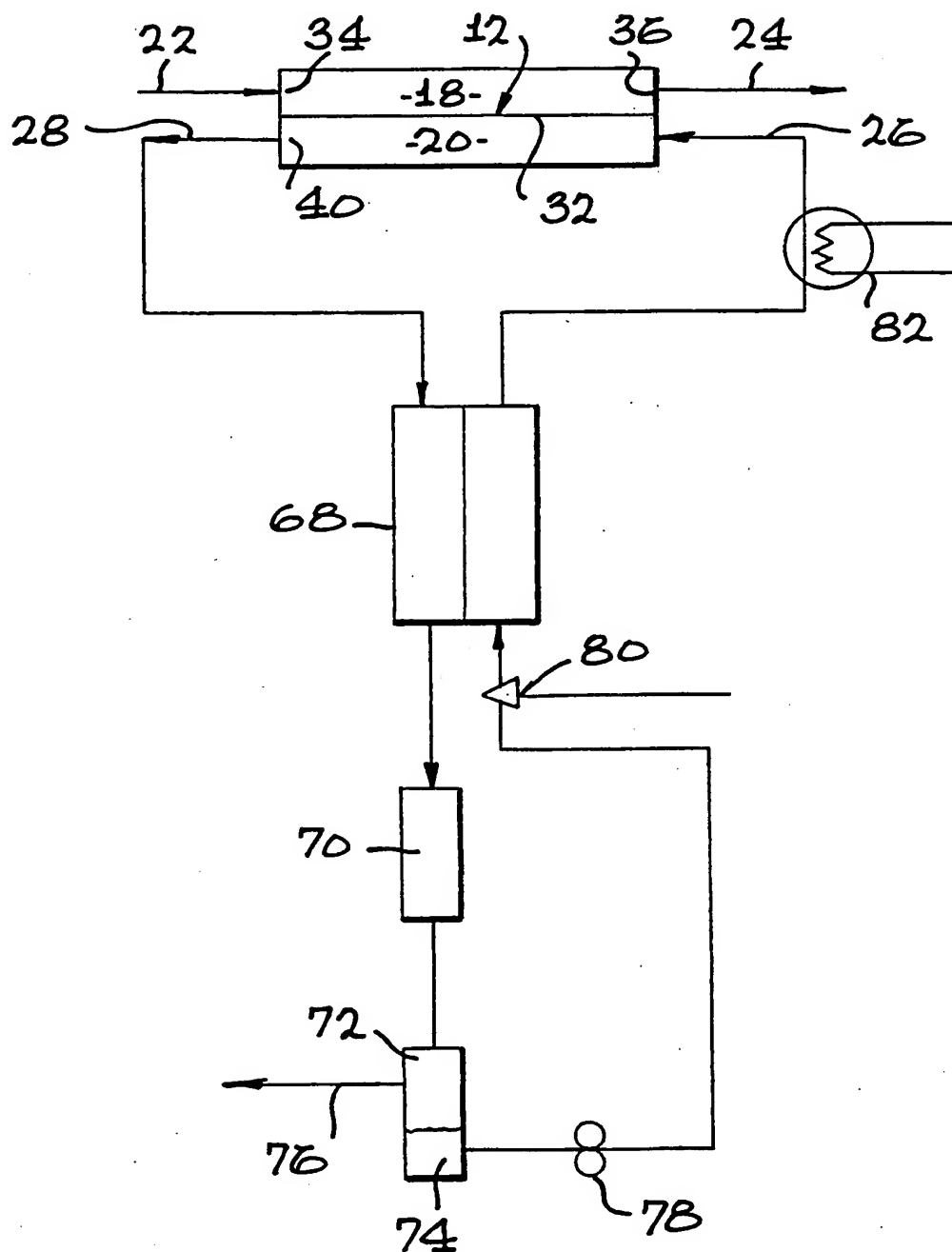
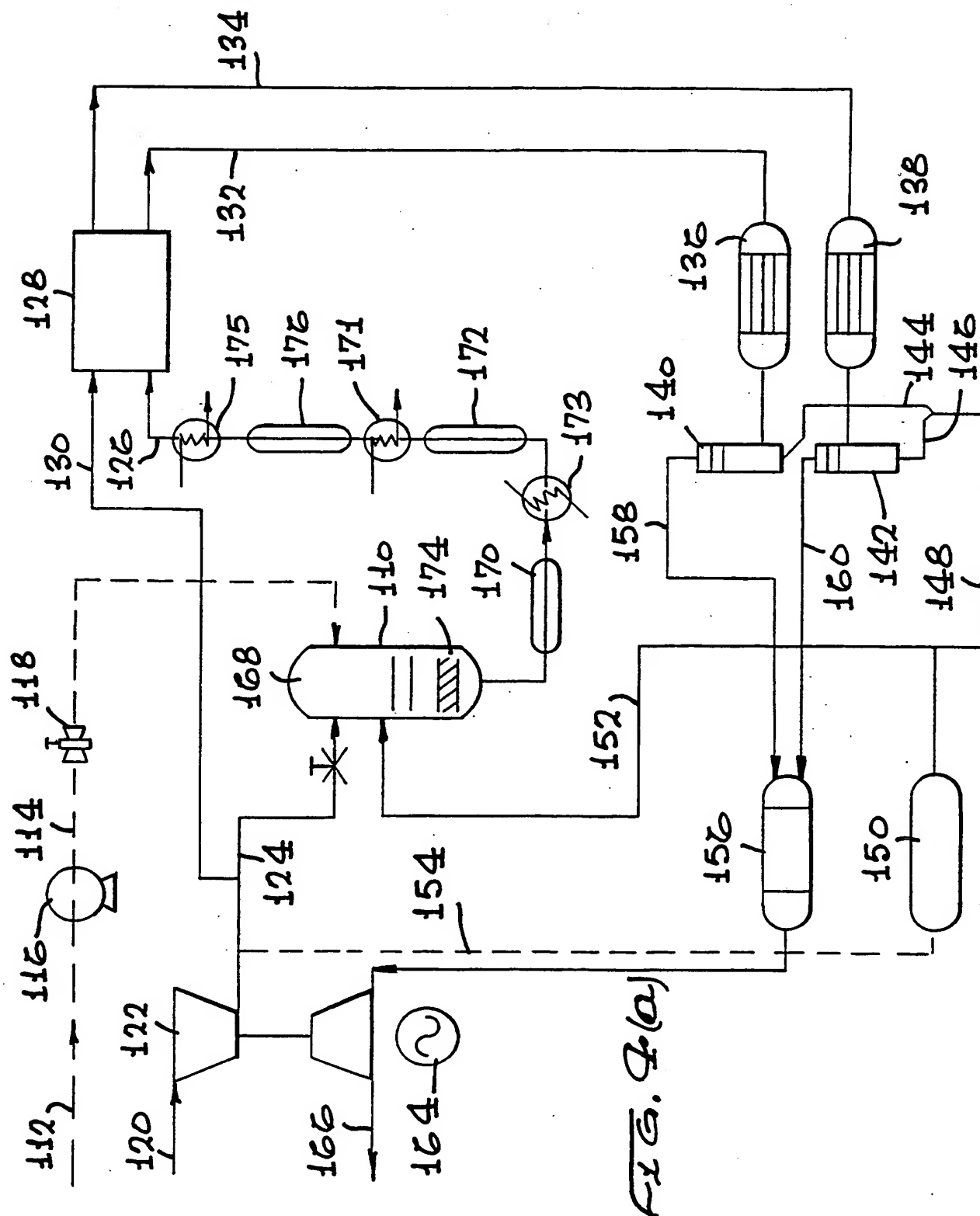


FIG. 4(b)

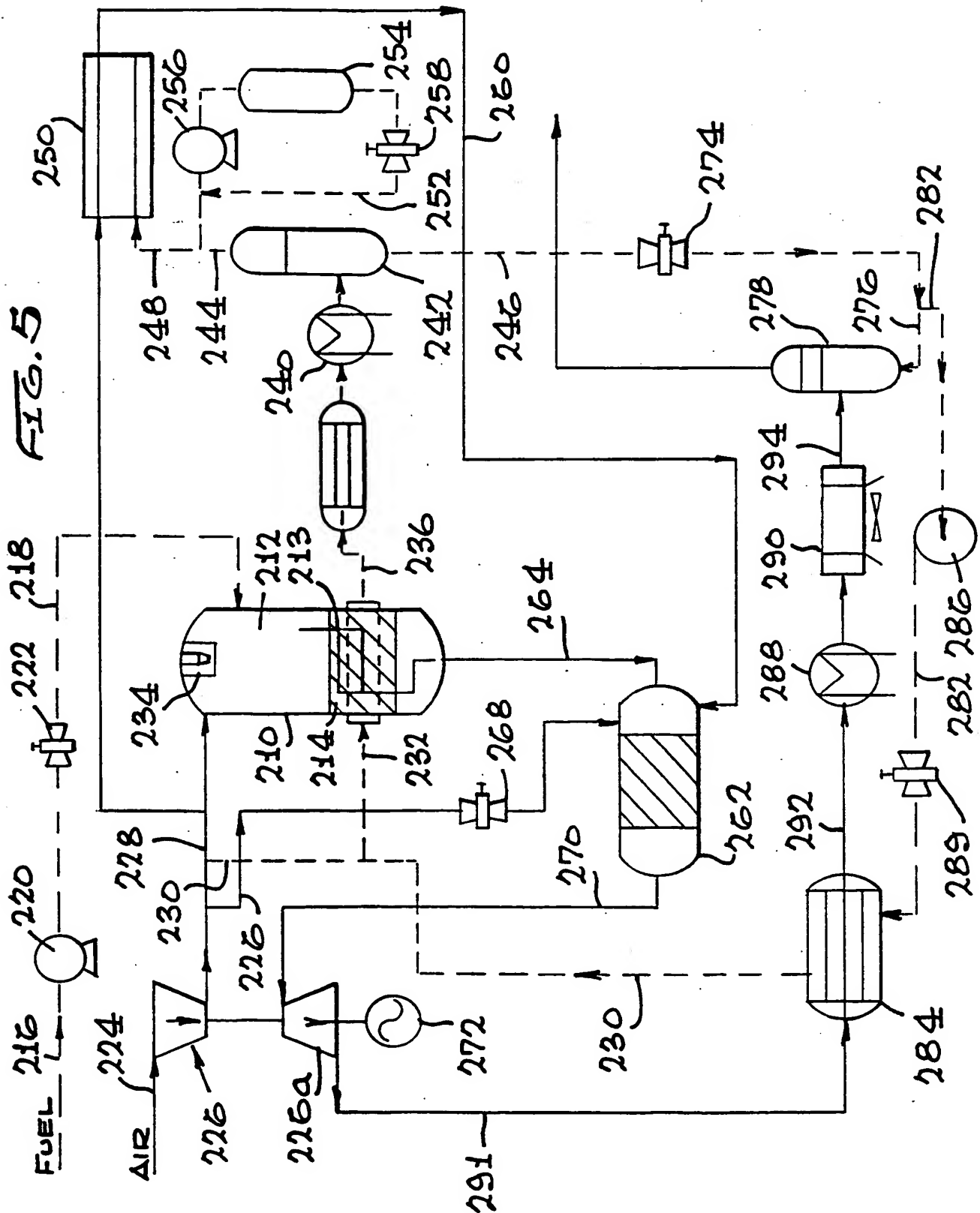
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FIG. 3





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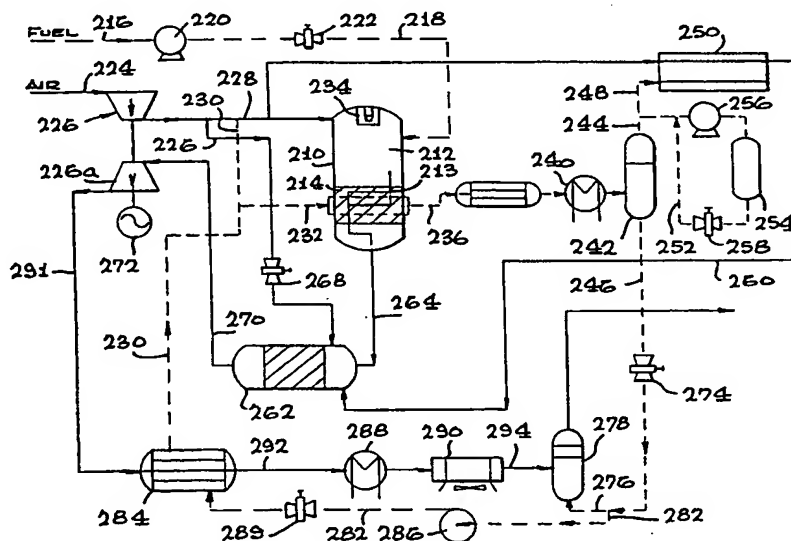




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(54) Title: PROCESS GAS PURIFICATION AND FUEL CELL SYSTEM



(57) Abstract

A module (214 Fig. 5) for separating a product from a mixed stream comprises a mixed stream chamber having inlet and outlet means and defining a first flow path for the mixed stream, a purge/product stream chamber having inlet and outlet means and defining a second flow path for a purge/product stream, the second flow path having a substantially countercurrent direction to that of the first flow path, and a membrane located between the mixed stream chamber and the purge/product stream chamber, the membrane being selectively permeable to the product. There is also disclosed a fuel cell system comprising a burner module (210) for mixing and combusting a fuel and air mixture to produce hydrogen rich fuel stream; a hydrogen fuel cell (250) for producing power/energy using the hydrogen fuel produced by the burner module; a hydrogen purification module (214) between the burner module and the fuel cell for extracting hydrogen fuel from the burner module for use in the fuel cell and that uses a purge gas to enhance purification module performance; hydrogen storage means (254) for storing hydrogen fuel produced by the burner module and not immediately required by the fuel cell; and means for feeding stored hydrogen fuel from the storage means to the fuel cell when the hydrogen requirements of the fuel cell are greater than the amount of hydrogen produced in the burner module.

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## INTERNATIONAL SEARCH REPORT

Internat Application No

PCT/US 99/05238

A. CLASSIFICATION OF SUBJECT MATTER		
IPC 6	B01D53/22	B01D53/32 H01M8/06 C01B3/50
According to International Patent Classification (IPC) or to both national classification and IPC		
B. FIELDS SEARCHED		
Minimum documentation searched (classification system followed by classification symbols)		
IPC 6 B01D H01M C01B		
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched		
Electronic data base consulted during the international search (name of data base and, where practical, search terms used)		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 562 754 A (KANG DOOHEE ET AL) 8 October 1996 (1996-10-08)  column 7, line 44 -column 9, line 41; figure 2  ---	1-4,6, 18-21, 25,28
X	EP 0 615 949 A (TOKYO GAS CO LTD ;MITSUBISHI HEAVY IND LTD (JP)) 21 September 1994 (1994-09-21) the whole document  ---	1-4,6, 18,20, 21,25,28
A	GB 990 131 A (ENGELHARD INDUSTRIES INC.) 28 April 1965 (1965-04-28) page 1, column 1, line 11 - line 33 page 2, line 4 - line 8 page 3, line 101 - line 109  ---  -/--	1-3,20, 21,28
<input checked="" type="checkbox"/> Further documents are listed in the continuation of box C. <input checked="" type="checkbox"/> Patent family members are listed in annex.		
* Special categories of cited documents : "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier document but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art. "&" document member of the same patent family		
Date of the actual completion of the international search		Date of mailing of the international search report
8 July 1999		26. 11. 99
Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016		Authorized officer  Borello, E

# INTERNATIONAL SEARCH REPORT

International Application No  
PCT/US 99/05238

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5 612 012 A (SOMA TAKAO ET AL) 18 March 1997 (1997-03-18) abstract; example 2 ---	1,20
A	GB 2 283 235 A (ROLLS ROYCE & ASS) 3 May 1995 (1995-05-03) abstract; figure 1 -----	1,20

# INTERNATIONAL SEARCH REPORT

International application No.  
PCT/US 99/05238

## Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:  
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☐ Claims Nos.:  
because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:
3. ☐ Claims Nos.:  
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

## Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☒ No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

1-4, 6, 18, 19-21, 25, 28, 29

### Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.
- ☐ No protest accompanied the payment of additional search fees.

## FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

## 1. Claims: 1-4,6,18,19-21,25,28,29

"A module and method for the purification of a product using a membrane and a condensable gas purge at the permeate side"

INDEPENDENT CLAIM 1 and its  
DEPENDENT CLAIMS: 2,3,4,6,18,19  
INDEPENDENT CLAIM 20 and its  
DEPENDENT CLAIMS: 21,25,28,29

Problem to be solved:

To purify a product with a membrane, using a sweep medium on the permeate side and to separate it from the product.

Solution:

To use a condensable gas

## 2. Claims: 1,5

"A module and method for the purification of hydrogen using the juxtaposition of a membrane with permeate side purging and a partial oxidation reformer as the source for the hydrogen containing mixed gas"

INDEPENDENT CLAIM 1 and its  
DEPENDENT CLAIM: 5

Problem to be solved:

To provide a mixed gas to produce purified hydrogen.

Solution:

To use a partial oxidation reformer.

## 3. Claims: 1,7,8,20,21,26,27,30

"A module and method for the purification of hydrogen using a diffusion membrane with permeate side purging"

INDEPENDENT CLAIM 1 and its  
DEPENDENT CLAIMS: 7,8  
INDEPENDENT CLAIM 20 and its  
DEPENDENT CLAIMS: 21,26,27,30 (as far as referred to the diffusion membrane)

Problem to be solved:

To separate the hydrogen from a gas mixture.

Solution:

To use a hydrogen diffusion membrane.

## 4. Claims: 1,9-13,20,21,23,24,30,45,46

## FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

"A module and method for the purification of hydrogen using a ion conducting membrane with permeate side purging"

INDEPENDENT CLAIM 1 and its

DEPENDENT CLAIMS: 9 (partially as far as referring to the ion conducting membrane), 10,11,12,13

INDEPENDENT CLAIM 20 and its

DEPENDENT CLAIMS: 21,23,24,30 ( partially as far as referred to the ion conducting membrane)

INDEPENDENT CLAIM 45: (partially: as far as referred to the method using a ion conducting membrane)

DEPENDENT CLAIM 46

Problem to be solved:

To separate the hydrogen from a gas mixture.

Solution:

To use a ion conducting membrane.

5. Claims: 1,9,14,15,20,21,23,24,30,45,46

"A module and method for the purification of hydrogen using a mixed ion and electron conducting membrane with permeate side purging"

INDEPENDENT CLAIM 1 and its

DEPENDENT CLAIMS: 9 (partially as far as referring to the mixed ion and electron conducting membrane), 14,15

INDEPENDENT CLAIM 20 and its

DEPENDENT CLAIMS: 21,23,24,30 ( partially as far as referred to the mixed ion and electron conducting membrane)

INDEPENDENT CLAIM 45: (partially: as far as referred to the method using a mixed ion and electron conducting membrane)

DEPENDENT CLAIM 46

Problem to be solved:

To separate the hydrogen from a gas mixture.

Solution:

To use a mixed ion and electron conducting membrane.

6. Claims: 1,16,17

"A module and method for the purification of a product using a membrane with permeate side purging and a catalyst for the CO shift reaction"

INDEPENDENT CLAIM 1 and its

DEPENDENT CLAIMS: 16,17

Problem to be solved:

**FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210**

To separate the product from a gas mixture and increase its concentration.

Solution:

To use a catalyst suitable for the CO shift reaction.

**7. Claims: 20,22**

"A module and method for the purification of a product using a membrane with permeate side purging and a higher pressure on the permeate side"

INDEPENDENT CLAIM 20

DEPENDENT CLAIMS: 22

Problem to be solved:

The tightness and the seal of the membrane to prevent the contamination of the high purity permeate gas by leakage from the feed/retentate side.

Solution:

To operate the permeate chamber at a higher pressure than the feed/retentate chamber.

**8. Claims: 31-44**

"A fuel cell system comprising: burner, hydrogen fuel cell, hydrogen purification module with permeate side purging"

INDEPENDENT CLAIM 31 (Device)

DEPENDENT CLAIMS: 32-38

INDEPENDENT CLAIM 39 (Process)

DEPENDENT CLAIMS: 40-44

Problem to be solved:

To set up and operate an integrated fuel cell system.

Solution:

To use a burner, a hydrogen purification module and a fuel cell.



# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 99/05238

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EP 0615949 A	21-09-1994	JP 6263402 A	20-09-1994
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